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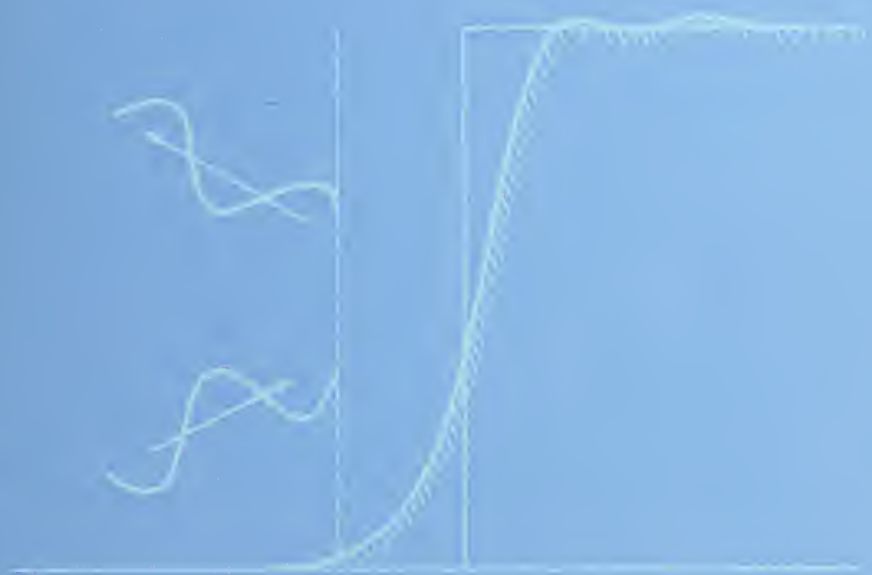
Metrological Consequences of the Hard Optical Boundary Assumption



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Metrological Consequences of the Hard Optical Boundary Assumption

A. George Lieberman

Center for Manufacturing Engineering
National Engineering Laboratory
National Bureau of Standards
Gaithersburg, MD 20899

U.S. DEPARTMENT OF COMMERCE, Malcolm Baldrige, Secretary
NATIONAL BUREAU OF STANDARDS, Ernest Ambler, Director

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Metrological Consequences of the Hard Optical Boundary Assumption

A. George Lieberman
National Bureau of Standards
Washington, DC 20234

ABSTRACT

The reflection of s-polarized light propagating in vacuum by a metal surface is examined for two descriptions of the vacuum-metal interface: the exponential surface transition and the Lang-Kohn transition. Both models treat the metal as a lossless, non-magnetic jellium material, but differ in the spatial distributions of their constituent charges. The displacement of the optical surface relative to the mechanical surface caused by the transition is evaluated for each model. Computerized results are presented for the optical displacement and phase change upon reflection from a sodium surface for the theoretically superior Lang-Kohn model. The measurement errors which result from ignoring the vacuum-metal transition become more significant as the angle of incidence is increased.

Key Words: exponential transition; hard boundary assumption; jellium model; Lang-Kohn transition; metal surface; optical boundary; optical displacement error; optical metrology; reflection coefficient; Ricatti equation; surface transition; wave immittance

1. INTRODUCTION

A clean, optically polished surface is customarily regarded as a physically abrupt termination to an otherwise uniform medium. This attitude is predicated upon a poor understanding of surface microstructure and the convenient applicability of classical Fresnel theory. However the consequences of neglecting a surface transition, occupying approximately 0.5 nm at a vacuum-metal interface, may not be insignificant.

Corrections accounting for the presence of an oxide or contaminant film, or the skin-depth of the electromagnetic field at a metal surface, are routine metrological considerations. Nevertheless, the systematic error introduced by the surface transition is not accounted for. In order to estimate the magnitude of this error for an arbitary metal a detailed analysis is provided for an alkali metal for which an excellent theoretical model [5][1] exists.

This report commences with a review of Fresnel's theory of reflection from a homogeneous medium (fig. 1a). This establishes a basis for interpreting the effect of a surface transition and a formalism for locating the apparent plane of reflection (optical surface) in the presence of a transition. Two descriptions of the vacuum-metal transition are examined. The first is an empirical model in which the electron density exhibits an exponential variation across the interface (fig. 1b). Fresnel theory does not apply here and the reflection coefficient must be determined from first principles; namely, as an electromagnetic boundary value solution of Maxwell's field equations. Analysis of the exponential transition leads to an analytical expression for the phase shift upon

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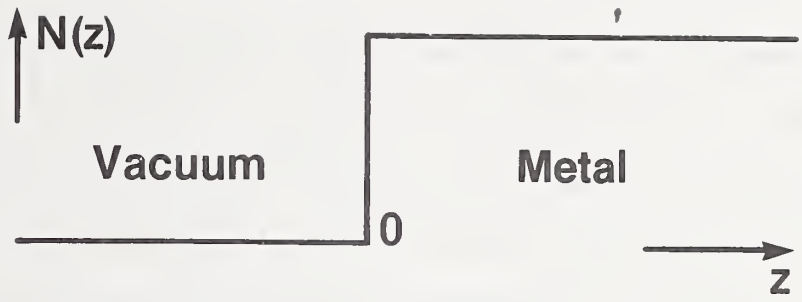
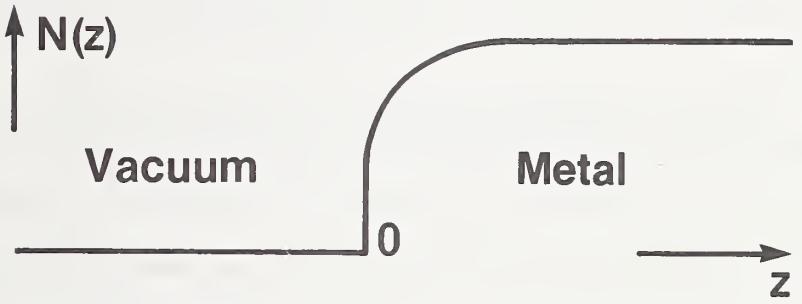
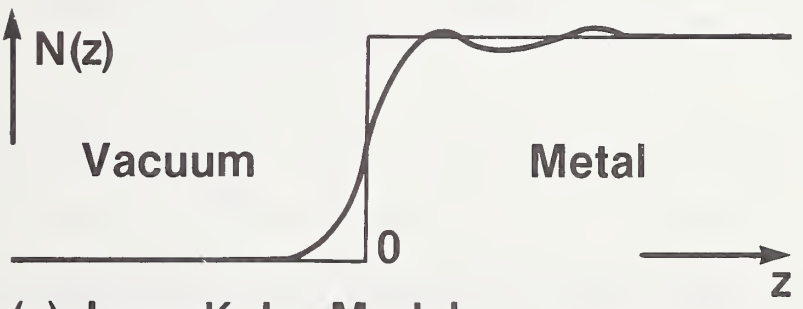
[1] Numbers in brackets indicate literature references listed at the end of the report.



(a) Hard Boundary Approximation



(b) Exponential Surface Transition



(c) Lang-Kohn Model

Figure 1. Three descriptions of the vacuum-metal interface. The electron and ion density distributions, N(z), are identical in (a) and (b) but not in (c) where the electron density distribution deviates from the uniform ion density.

reflection, and locates the optical surface behind the mechanical surface of the metal.

The second description of the vacuum-metal interface is a semi-classical theoretical model known as the Lang-Kohn transition (fig. 1c). The electron density is specified numerically but fortunately knowledge of the field configuration is not essential for determining the phase shift of the reflected field. Instead, a formulation based on the wave admittance is used to calculate the phase shift. In this case the apparent plane of reflection is found to be positioned slightly in front of the ion surface.

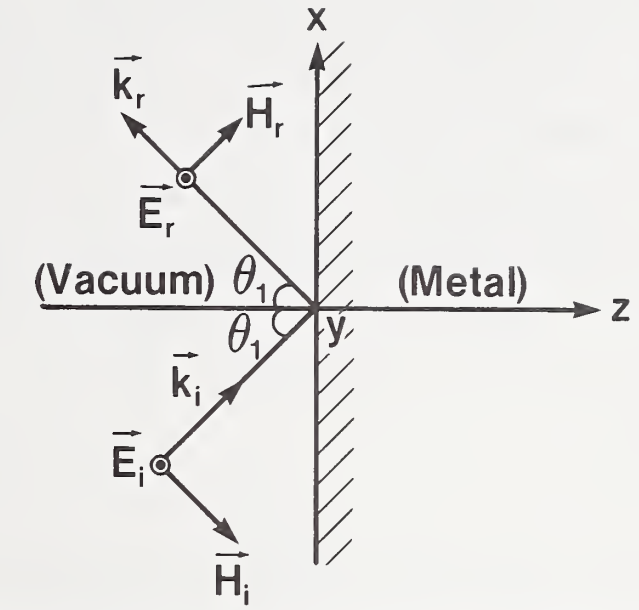
2. THE HARD OPTICAL BOUNDARY AND FRESNEL THEORY

Consider the reflection into vacuum of an s-polarized monochromatic plane wave from the surface of an arbitrary depth-dependent medium (fig. 2). The total electric field in the vacuum region ($z < 0$) due to the incident wave and its reflection is given by the real part of

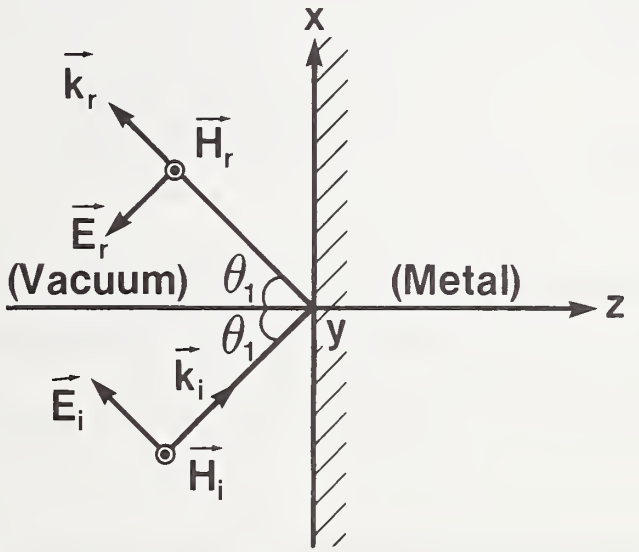
$$\begin{aligned}\vec{E} &= \vec{E}_i\, e^{-i(\omega t - k_x x)}\, [e^{ik_z z} + R\, e^{-ik_z z}] \\ &= \vec{E}_i\, e^{-i(\omega t - k_x x - k_z z)}\, [1 + |R| e^{i(\phi_o - 2k_z z)}],\end{aligned} \tag{2-1}$$

where $R = |R| e^{i\phi_o}$ is the reflection coefficient of the half-space evaluated, for any time t, at the boundary $z = 0$. At this plane, R is the ratio of the complex vector field amplitude of the reflected wave to that of the incident wave, $\vec{E}_i$. The total intensity, I, of the radiation within this region is proportional to $|E|^2$. Thus

$$I = I_i[1 + |R|^2 + 2|R|\cos(\phi_o - 2k_z z)], \tag{2-2}$$



(a) Reflection of an s-Polarized Plane Wave



(b) Reflection of a p-Polarized Plane Wave

Figure 2. Field directions for the two principal polarizations of a plane wave. (a) Reflection of an s-polarized plane wave. (b) Reflection of a p-polarized plane wave.

where I_i represents the intensity of the incident light. Analogous expressions pertain to the magnetic field of a p-polarized plane wave.

Equation (2-2) shows that the intensity of the total field is independent of time and consists of a constant term plus a periodic function of position. The intensity oscillates between a maximum value $I_{max} = I_i(1+|R|)^2$ and a minimum value of $I_{min} = I_i(1-|R|)^2$ with spatial periodicity $\Delta z = 2\pi/2k_z = (1/2)\lambda\cos\theta_1$, where θ_1 is the angle of incidence.

The magnitude and phase of R are determined by the polarization and incidence angle of the incoming wave, and by the depth-dependence of the material parameters beneath the surface. In any particular case both $|R|$ and ϕ_o are fixed. By definition ϕ_o is the phase shift in the field upon reflection from the plane z = 0. At some other plane located at z = -d, eq. (2-2) shows that the effective phase shift is

$$\phi(d) = \phi_o + 2k_z d \quad . \tag{2-3}$$

Therefore, as a consequence of the periodicity of the fields, it is also possible to regard the reflection as having effectively occurred at the plane z = -d provided only that the phase shift ϕ_o is increased by $2k_z d$.

The reflection and transmission coefficients are usually determined by matching the field solutions of Maxwell's equations across the vacuum-material interface. For a passive, linear, isotropic medium whose constitutive parameters vary only with depth below the surface, field continuity across the interface ensures that: reflection occurs without change of frequency; the propagation directions of the reflected and

refracted waves do not depend upon the incident wave polarization; and a linearly polarized plane wave retains its polarization upon reflection. Field continuity further requires an equality of the angles of incidence and reflection, and also that the angle of refraction, $\theta_t(z)$, of the transmitted wave at depth z where the refractive index is $\eta(z)$, satisfies Snell's law:[2]

$$\eta(z)\sin\theta_t(z) = \sin\theta_1. \tag{2-4}$$

If, in addition, the surface transition can be neglected (the hard optical boundary approximation) then field continuity also leads to the following Fresnel relations for the reflection coefficient, R, and the Fresnel transmission coefficient, T, for an s-polarized plane wave [1]:

$$R = \frac{E_r}{E_i} = -\frac{1 - \sqrt{1+(\eta^2-1)/\cos^2\theta_1}}{1 + \sqrt{1+(\eta^2-1)/\cos^2\theta_1}} = 1 - T \tag{2-5}$$

The refractive index of a metal, characterized as a free-electron jellium of plasma frequency ω_p [2], is given by

$$\eta^2 = (1 - \omega_p^2/\omega^2) \quad , \text{ where } \quad \omega_p^2 = Ne^2/m\varepsilon_o. \tag{2-6}$$

---

[2] The validity of the law of reflection at the surface of a graded material an be argued on the physical grounds that: (1) the optical path is invariant under interchange of source and receiver (Helmholtz reciprocity theorem) and (2) the medium is invariant under a 180° rotation about the surface normal. The validity of Snell's law for describing the continuous refraction of the optical path within a graded medium follows upon dividing the medium into infinitesimally-thin, uniform slabs parallel to the surface and applying Snell's law to each interface. A rigorous theoretical proof is given in reference [4].

N is the electron density, and m is the mass of an electron. The plasma frequency of a metal is several times larger than the frequency of visible light, so it is appropriate to express R as the complex number:

$$R = \frac{1 - i\sqrt{(\omega_p'/\omega)^2 - 1}}{1 + i\sqrt{(\omega_p'/\omega)^2 - 1}} = \exp(i\phi_o), \tag{2-7}$$

where $\omega_p' = \omega_p/\cos\theta_1$. The incident s-polarized plane wave is totally reflected ($|R| = 1$) for $\omega < \omega_p'$ and the phase shift upon reflection at the vacuum-metal interface is

$$\phi_o = -2\tan^{-1}\sqrt{(\omega_p'/\omega)^2 - 1}\ . \tag{2-8}$$

In accordance with eq. (2-3) at any other position, located at a distance d in front of the surface, the phase shift is

$$\phi(d) = 2k_z d - 2\tan^{-1}\sqrt{(\omega_p'/\omega)^2 - 1}\ . \tag{2-9}$$

Consider, as an example, the reflection of a He-Ne laser beam ($\lambda = 6.330\times10^{-7}$ m, $\omega = 2\pi c/\lambda = 2.97781 \times 10^{15}$rad/s) by a sodium surface. If the metal is treated as homogeneous body of electron density $N = 2.5 \times 10^{28}m^3$ ($\omega_p = 8.91973 \times 10^{15}$rad/s) and having a zero-width surface transition, then the phase shift upon reflection for normal incidence is $\phi_o = -140.995°$. By comparison, the phase shift upon reflection from a perfect conductor would be $\phi_o = -180°$ corresponding to infinite ω_p and zero skin depth. For a good conductor such as aluminum, the plasma frequency is $\omega_p = 6.07889 \times 10^{16}$rad/sec and $\phi_o = -174.384°$.

The Fresnel coefficients are inappropriate for graded materials because the waves that are able to propagate within an inhomogeneous medium are not planar. However, an approximate treatment utilizing Fresnel theory can be given by dividing the inhomogeneous medium into homogeneous strata which do support plane waves. As a result of this discretization, a characteristic matrix can be defined for each individual stratum and then for the whole system [3]. This formulation readily lends itself to numerical analysis but requires considerable judgement in defining the strata. Another approach has been to solve Maxwell's equations for some idealized material profile and then to identify the reflection coefficient with the ratio of the appropriate "outgoing" to "incoming" field amplitudes at some exterior position where the fields resume the character of plane waves.

The remainder of the report examines the reflection of plane waves from metal surfaces characterized by non-zero transition widths. The transition is initially modeled as having an exponential depth profile and then, more accurately, as having a Lang-Kohn profile.

3. THE EXPONENTIAL SURFACE TRANSITION

The Fresnel relations apply strictly to the reflection and transmission of plane waves at a plane interface between two homogeneous media. At such an interface the material properties are assumed to change abruptly from their constant values on either side. On a microscopic level this idealization, at the very least, violates thermodynamic equilibrium by ignoring such fundamental processes as interdiffusion, or for a free surface: restructuring of the surface to accomodate severed chemical bonds, impurity absorption, and surface oxidation. For many macroscopic observations these processes may be neglected and the Fresnel relations suffice. However, as instrumentation

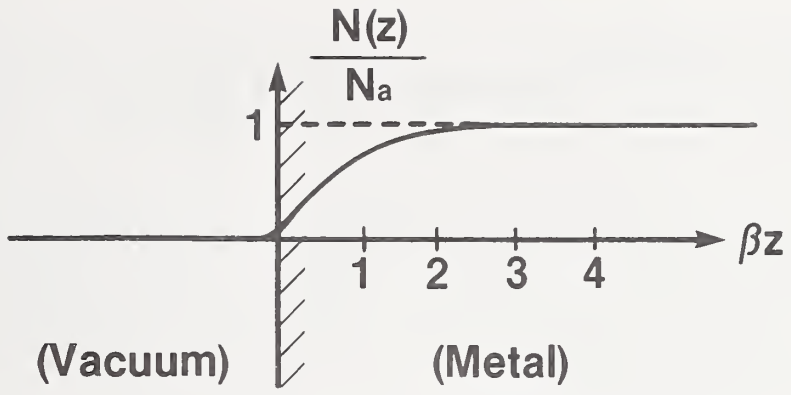
and techniques improve, the existence of a surface transition becomes increasingly difficult to ignore. It is, therefore, relevant to examine the effect of such a transition in the optical properties of the metal. Consider a metal which occupies the half-space $z \geq 0$ and is modeled by a jellium permittivity function (fig. 3) which decreases exponentially with depth from the surface:

$$\varepsilon(z) = \begin{cases} \varepsilon_o, & z < 0. \\ \varepsilon_o[1 - (\omega_p/\omega)^2 (1 - e^{-\beta z})], & z \geq 0. \end{cases} \tag{3-1}$$

The plasma frequency deep within the interior, ω_p, and the characteristic length of the transition, $1/\beta$, are constants of the medium. At the plane $z=0$ the permittivity is equal to the free-space dielectric constant ε_o, while at great depths within the metal (measured in units of $1/\beta$) the permittivity is reduced to the usual bulk value $\varepsilon_o[1-(\omega_p/\omega)^2]$.

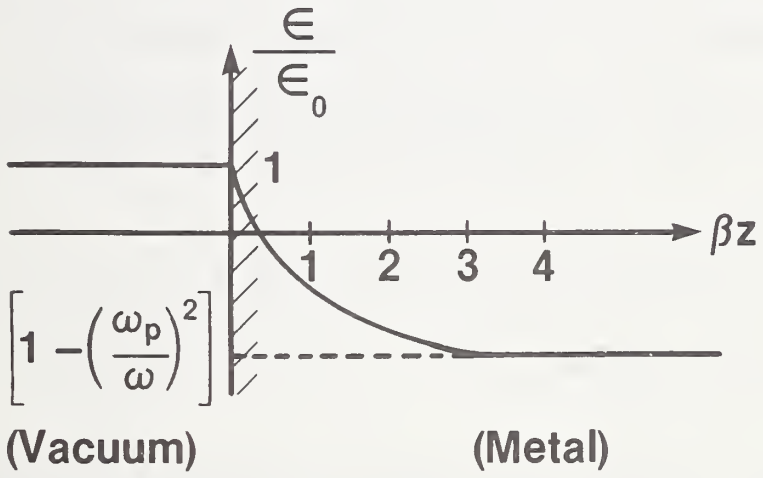
Suppose this medium is illuminated by an s-polarized monochromatic plane wave, incident at angle θ_1, with its electric vector entirely in the y direction (fig. 2a). As for a homogeneous medium the incident, reflected, and transmitted fields may each be expressed as the real part of a complex vector function containing as factors an $\exp(-i\omega t)$ temporal dependence and an $\exp(ik_o \sin\theta_1 x)$ transverse spatial dependence. For the sake of notational brevity these factors are suppressed and the fields of the incident plane wave are simply:

$$\vec{E}_i = E_o e^{+ik_o \cos\theta_1 z} \hat{e}_y \quad , \tag{3-2}$$



(a) Electron Density Profile:

$$\frac{N(z)}{N_a} = \left(1 - e^{-\beta z}\right), \; z \geq 0.$$



(b) Dielectric Permittivity Profile:

$$\frac{\in}{\in_0} = \left[1 - \left(\frac{\omega_p}{\omega}\right)^2 \left(1 - e^{-\beta z}\right)\right], \; z \geq 0.$$

Figure 3. The exponential surface transition. (a) The electron density profile. (b) The dielectric permittivity profile.

$$\vec{H}_i = \sqrt{\frac{\varepsilon_o}{\mu_o}}\, E_o\, e^{+ik_o \cos\theta_1 z}[-\cos\theta_1 \hat{e}_x + \sin\theta_1 \hat{e}_z] \quad , \tag{3-3}$$

where E_o is a complex scalar constant, and (e_x, e_y, e_z) is the triad of unit vectors along the three coordinate directions. The reflected plane wave fields are likewise written:

$$\vec{E}_r = R\, E_o\, e^{-ik_o \cos\theta_1 z}\, \hat{e}_y \quad , \tag{3-4}$$

$$\vec{H}_r = R \sqrt{\frac{\varepsilon_o}{\mu_o}}\, E_o\, e^{-ik_o \cos\theta_1 z}[\cos\theta_1 \hat{e}_x + \sin\theta_1 \hat{e}_z] \quad . \tag{3-5}$$

In order to emphasize the as yet undetermined z dependence, the fields of the transmitted wave in the inhomogeneous metal are expressed as:

$$\vec{E}_t = E_y(z)\, \hat{e}_y \quad , \tag{3-6}$$

$$\vec{H}_t = [H_x(z)\hat{e}_x + H_z(z)\hat{e}_z] \quad . \tag{3-7}$$

When due account is taken of the implicit dependence of the fields on x and t the two Maxwell equations for a non-permeable medium ($\mu = \mu_o$)

$$\nabla \times \vec{E} = -\mu_o \frac{\partial \vec{H}}{\partial t} \quad , \tag{3-8}$$

$$\nabla \times \vec{H} = \varepsilon \frac{\partial \vec{E}}{\partial t} \quad , \tag{3-9}$$

upon substitution of eqs. (3-6) and (3-7) for the transmitted fields and

eq. (3-1) for the permittivity function, become

$$-\frac{dE_y}{dz} = i\omega\mu_o H_x \quad , \tag{3-10}$$

$$ik_x E_y = i\omega\mu_o H_z \quad , \tag{3-11}$$

$$\frac{dH_x}{dz} - ik_x H_z = -i\omega\varepsilon E_y \quad , \tag{3-12}$$

where $k_x = k_o \sin\theta_1$. Elimination of H_x and H_z from eq. (3-12) by means of eqs. (3-10) and (3-11) leads to the following differential equation for the transmitted electric field amplitude:

$$\frac{d^2E_y}{dz^2} + k_o^2 \left[\cos^2\theta_1 - (1 - \omega_p^2/\omega^2)e^{-\beta z}\right]E_y = 0 \quad . \tag{3-13}$$

To facilitate solution, z is replaced by a new dimensionless variable v defined by the transformation

$$v = 2\frac{\omega_p}{\omega}\frac{k_o}{\beta}e^{-\beta z/2} \quad . \tag{3-14}$$

The electric field expressed as a function of v now satisfies Bessel's differential equation,

$$\frac{d^2E_y}{dv^2} + \frac{1}{v}\frac{dE_y}{dv} + \left[1 - (\nu/v)^2\right]E_y = 0 \quad , \tag{3-15}$$

with

$$\nu = 2(k_o/\beta)\sqrt{(\omega_p'/\omega)^2 - 1} \qquad (3\text{-}16)$$

and $\omega_p' = \omega_p/\cos\theta_1$. The formal solutions of the transformed differential equation are linear combinations of $J_{\pm\nu}(v)$; for small arguments $J_{\pm\nu}(v)$ behaves as $v^{\pm\nu}$. Index ν is real according to (3-16) because the frequency of visible light is always smaller than the reduced plasma frequency ω_p' of the metal. For the fields to remain finite at very large z (small v), only Bessel functions of positive order are permitted in the solution of eq. (3-15). Therefore

$$E_y(v) = C_\nu J_\nu(v), \qquad (3\text{-}17)$$

where C_ν is a constant proportional to the amplitude of the incident wave. The magnetic field component parallel to the surface plane is evaluated by substituting $E_y(v)$ into Maxwell's equation and obtaining

$$H_x(v) = \frac{-i\beta v}{2\omega\mu_o} C_\nu J_\nu'(v) \quad , \qquad (3\text{-}18)$$

where $J_\nu'(v)$ denotes the derivative of Bessel function $J_\nu(v)$ with respect to its argument v. Matching the total (tangential) electric fields of each half-space across the surface z = 0 ($v = v_o = 2\,\omega_p k_o/\omega\beta$), and repeating this for the tangential components of the total magnetic fields results in two equations in the two unknowns C_ν and R. Eliminating C_ν leads to the following expression for the reflection coefficient at the jellium surface:

$$R^{exp}(0) = \frac{1 - i(\omega_p'/\omega)\, J_\nu'(v_o)/J_\nu(v_o)}{1 + i(\omega_p'/\omega)\, J_\nu'(v_o)/J_\nu(v_o)} \quad . \tag{3-19}$$

The phase shift in the electric field upon its reflection at the surface $z = 0$ is given by

$$\phi_o{}^{exp} = \arg R^{exp} = -2\tan^{-1}\left[(\omega_p'/\omega)J_\nu'(v_o)/J_\nu(v_o)\right] \tag{3-20}$$

As a verification of the solution consider the result of passing to the limit, $\beta\to\infty$. The exponential profile function (3-1) reduces to the step profile function of a homogeneous half-space, and in this limit $v_o\to 0$, $J_\nu(v_o) \to v_o{}^\nu$, and

$$R^{exp}(0) \to \frac{1 - i\ (\omega_p'/\omega)(\nu/v_o)}{1 + i\ (\omega_p'/\omega)(\nu/v_o)} \tag{3-21}$$

wherein

$$\nu/v_o = \sqrt{(\omega_p'/\omega)^2 - 1}\ \Big/\ (\omega_p'/\omega) \quad . \tag{3-22}$$

As required, the last two expressions combine to yield the usual Fresnel reflection coefficient, cf. eqs. (2-7) and (2-8), for an s-polarized plane wave

$$R^{exp}(0) \to \frac{1 - i\sqrt{(\omega_p'/\omega)^2 - 1}}{1 + i\sqrt{(\omega_p'/\omega)^2 - 1}} \quad . \tag{3-23}$$

The difference between the phase shift for an s-polarized plane wave reflected from an exponential transition and that from a step

transition, both located at z = 0 and both having the same interior density, is given by

$$\Delta\phi^{exp} = \phi_o{}^{exp} - \phi_o{}^{step}$$
$$= 2\left\{\tan^{-1}\sqrt{(\omega_p'/\omega)^2 - 1} - \tan^{-1}[(\omega_p'/\omega)\, J_\nu'(v_o)/J_\nu(v_o)]\right\} \qquad (3\text{-}24)$$

Consider the magnitude of this quantity for a sodium surface illuminated by a normally incident He-Ne laser beam. Taking $1/\beta = 0.2$ nm as the characteristic length of the material transition, then by direct computation: $v_o = 1.189300 \times 10^{-2}$, $\nu = 1.121068 \times 10^{-2}$, $J_\nu(v_o) = 9.501637 \times 10^{-1}$ and $J_\nu'(v_o) = 8.900633 \times 10^{-1}$. This leads to a relative phase shift $\Delta\phi^{exp} = 0.226°$. Since the time dependence of the fields is given by $\exp(-i\omega t)$, the positive value of $\Delta\phi^{exp}$ represents a relative delay. The phase delay caused by the exponential transition is consequently larger than that caused by a step transition of equal magnitude at z = 0 because, for the same interior density, the incident wave must penetrate the exponential transition to a greater depth before encountering a permittivity small enough to cause total reflection.

A more fundamental explanation of the relative phase delay rests with the jellium model. The optical reflection process is completely governed by the collective oscillations of the jellium electrons about the lattice sites of the fixed positive ions. The lattice of positive ions not only defines the mechanical shape of the solid but also provides charge neutrality at each location within the jellium. Local charge neutrality further implies that the electrons and the ions have identical spatial distributions. Both distributions are bounded by the same plane

which in this model, as in the Fresnel model, unambiguously locates the metal surface at z = 0. The lower density of electrons available for optical scattering in the exponential surface transition, as compared to a step transition of equal magnitude, results in a larger penetration path and phase shift for the totally reflected incident wave.

Although the effects of oxide and contaminant films, losses and skin depths are taken into consideration by the optical metrologist in order to correct the phase shift and account for the penetration of the optical field into a metal end standard, the surface transition itself is ignored as soon as the Fresnel reflection formulae are applied. By using the Fresnel formulae the metrologist implicitly defines the location of an apparent plane of reflection for the surface. The location of this apparent plane of reflection for the exponential transition is determined theoretically by setting the relative phase shift $\Delta\phi_s^{exp}$ calculated in (3-24) equal to $2k_z\delta$ in accordance with eq. (2-3). Thus

$$\Delta\phi^{exp} = \phi_o{}^{exp} - \phi_o{}^{step} = 2k_z\delta^{exp} \tag{3-25}$$

or, equivalently,

$$\delta^{exp} = \frac{1}{k_z}\left\{ \tan^{-1}\sqrt{(\omega_p'/\omega)^2 - 1} - \tan^{-1}\frac{\omega_p' J_\nu'(v_o)}{\omega J_\nu(v_o)} \right\} . \tag{3-26}$$

For the parameters of the preceeding example, the apparent plane of reflection (i.e. optical surface) is located at the distance δ^{exp} = 0.3974 nm below the vacuum-metal interface. Because of local charge neutrality, δ^{exp} has the added significance of being equal to the

displacement of the optical surface from the mechanical surface.

The formulation presented next provides a method for calculating the reflection coefficient and phase shift without having to solve Maxwell's equations. It will prove particularly convenient for treating the Lang-Kohn model of a metal surface.

4. SUMMARY OF THE REFLECTION COEFFICIENT FORMULATION AND THE WAVE ADMITTANCE FORMULATION

In a companion paper [4] a differential formulation was described for the s-polarized plane wave reflection coefficient R(0) at the surface to an arbitrary vacuum-material transition. For present purposes the medium is a jellium metal having a specified, depth-dependent plasma frequency

$$\omega_p(z) = \sqrt{\frac{e^2}{m\varepsilon_o} N(z)} \tag{4-1}$$

where N(z) is the electron density profile. The reflection coefficient is determined by evaluating at the metal surface the solution R(z) of the differential equation:

$$\frac{dR}{dz} + i2\kappa(z)R - \Gamma_s(z)[1 - R^2] = 0 \tag{4-2}$$

This differential equation is non-linear, first order and has the form of a standard Ricatti equation. Solution R(z) is subject to the one boundary condition, viz.,

$$R(z_s) = +1 \tag{4-3}$$

The depth z_s, at which eq. (4-3) indicates total reflection occurs, is determined from:

$$\omega_p(z_s) = \omega\cos\theta_1 \quad . \tag{4-4}$$

The coefficients of differential equation (4-2) are given by:

$$\kappa(z) = \frac{1}{c}\sqrt{\omega^2\cos^2\theta_1 - \omega_p{}^2(z)} \quad , \tag{4-5}$$

and

$$\Gamma(z) = -\frac{1}{4}\,[\omega^2\cos^2\theta_1 - \omega_p{}^2(z)]^{-1}\,\frac{d}{dz}\,(\omega_p{}^2) \tag{4-6}$$

where $c = 1/\sqrt{\mu_o\varepsilon_o}$ is the vacuum speed of light. In solving eq. (4-2) cognizance must be taken of the singularity in $\Gamma(z)$ at z_s.

A less troublesome formulation makes use of the wave admittance Y(z) which at any depth z is related to R(z) via the bilinear transformation

$$R(z) = -\frac{Y(z) - \kappa(z)/\omega\mu_o}{Y(z) + \kappa(z)/\omega\mu_o} \quad . \tag{4-7}$$

The differential equation for Y(z) is obtained by substituting transformation (4-7) into eq. (4-2). As a result, Y(z) is found to satisfy the equation

$$\frac{dY}{dz} + i\omega\mu_o Y^2 - i\omega\varepsilon_o[\cos\theta_1 - \omega_p{}^2(z)/\omega^2] = 0 \tag{4-8}$$

Unlike eq. (4-2) the coefficients of this Ricatti-type equation are

nowhere singular. Applying transformation (4-7) to eq. (4-3) the boundary condition on Y(z), corresponding to total reflection at depth z_s, is obtained:

$$Y(z_s) = 0 \quad . \tag{4-9}$$

Either the R(z) formulation or the Y(z) formulation may be utilized to compute the reflection coefficient from the electron density distribution, without ever having to solve Maxwell's equations for the electromagnetic fields. This is especially advantageous when the electron density is numerically specified.

5. THE LANG-KOHN SURFACE TRANSITION

The Lang-Kohn model [5] is a semi-classical, theoretical description of an alkali metal surface. Although the applicability of this model to other metal surfaces is generally more empirical than theoretical in nature it is presently the best available model. In the Lang-Kohn model the metal lattice is characterized by a uniform distribution of positive ions which is abruptly terminated at the surface z = 0 separating it from the ambient vacuum (figs. 1c and 4). The electron distribution is permitted to spread into the vacuum beyond the ion surface creating, thereby, a local charge imbalance and a corresponding restoring potential. The equilibrium spatial distribution of the electrons is determined by the simultaneous solution of Schroedinger's equation and Poisson's equation. Because Schroedinger's equation for the electronic probability amplitude depends upon the electrostatic potential of the unneutralized charge density and the latter depends on the former, the electron distribution is computed by a means of self-consistent iterative numerical technique.

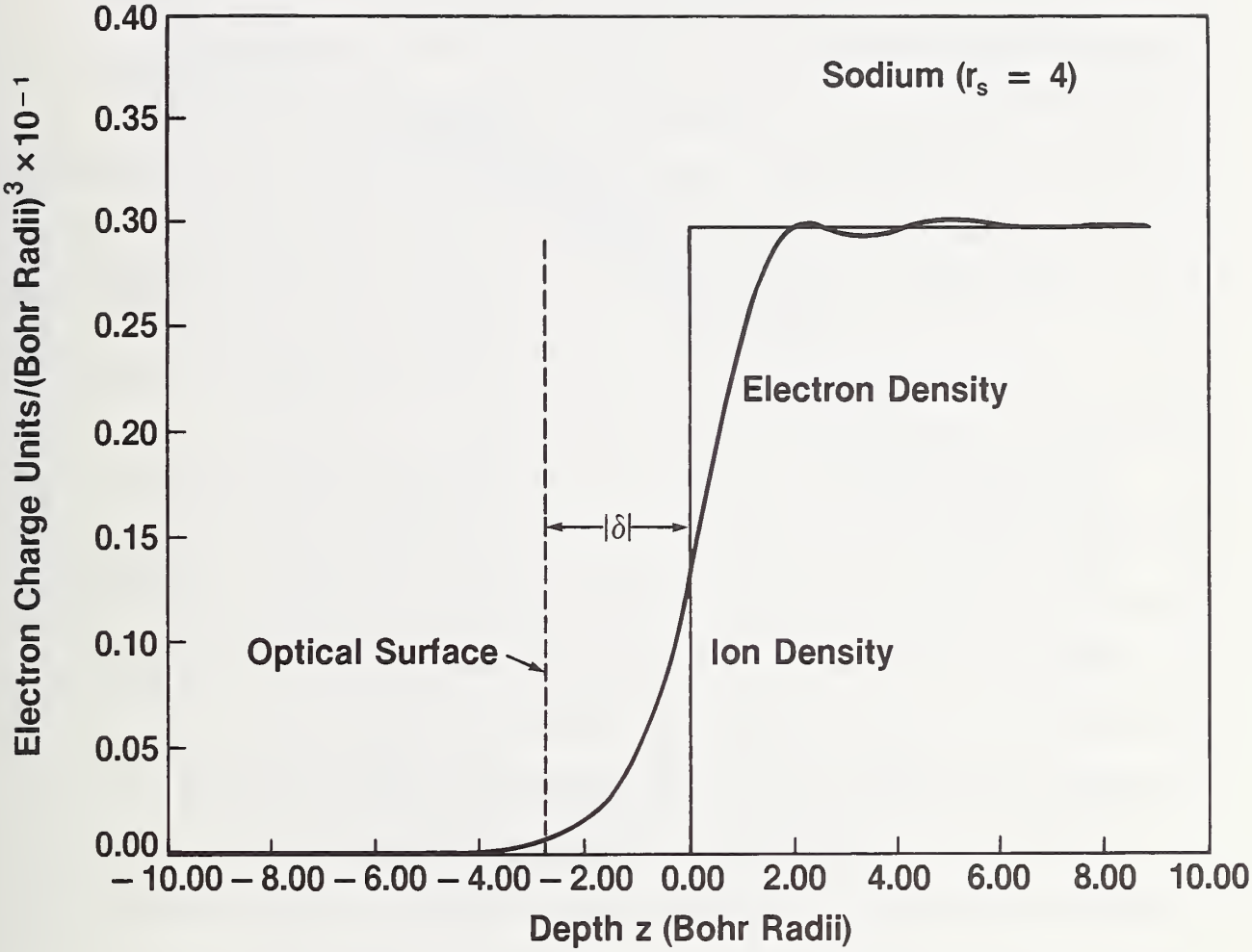


Figure 4. The electron and ion densities, and a typical location of the apparent plane of reflection (optical surface), for a vacuum-metal interface modeled by a Lang-Kohn transition. The electron density plot is based upon data provided in Table 1 of the Lang and Kohn paper [5].

Since the electron distribution, and consequently the permittivity of the metal surface, are provided by the Lang-Kohn model only in tabular numerical form, it is neither practical nor desirable to solve Maxwell's equations for the electromagnetic field configuration. The complex reflection coefficient is more expediently evaluated via the admittance formulation presented earlier. The admittance formulation is sufficiently general that any layered system can be investigated through its use. For example, with the appropriate modification of eqs. (4-5) and (4-6) for $\kappa(z)$ and $\Gamma(z)$ [4] the effects of oxide and contaminant films on optical reference surfaces can also be explored.

The electron and ion densities at a vacuum-metal interface are graphed in figure 4 from data provided in the original paper by Lang and Kohn [5]. The electron density profile presented in this figure corresponds to the metal sodium which has a Wigner-Seitz radius [6] $r_s = 4.006$. Notice that the density profiles are expressed in atomic units, i.e., all lengths (including r_s) are scaled in units of the Bohr radius $a_o = 0.0529572$ nm. Due to the extension of the electron distribution beyond the ion surface the apparent plane of reflection (optical surface) of the metal is displaced toward the vacuum by a distance $|\delta|$. The value of $|\delta| \simeq 3a_o$ shown in figure 4 is typical of incidence at 45° by a beam of visible light.

The computational results of applying the admittance formulation to the Lang-Kohn model are shown in figure 5. As earlier, $\Delta\phi$ represents the phase shift caused by the presence of the transition. In this case $\Delta\phi$ is calculated as the difference between the phase shift determined for the Lang-Kohn model and the phase shift determined for a step transition (i.e., hard boundary) corresponding the same interior electron density.

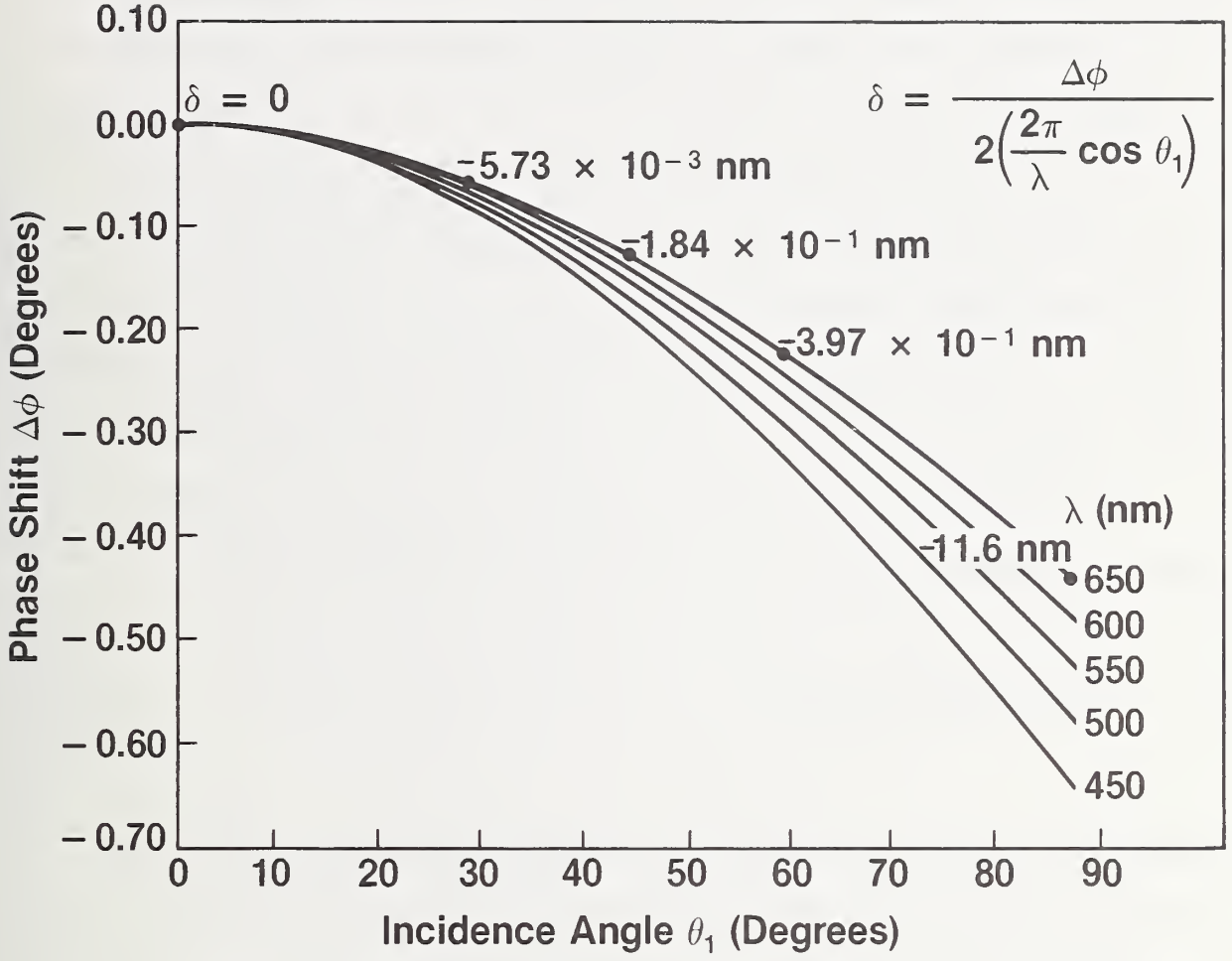


Figure 5. Relative phase shift, $\Delta\phi$, defined by eq. (5-1), and displacement of the optical surface, δ, as functions of the incidence angle, θ_1, and wavelength, λ, for the Lang-Kohn transition of Figure 4.

In both cases the ion surfaces are located at the plane z = 0. It follows that

$$\Delta\phi = \phi_d{}^{L-K} - \phi_d{}^{step} \quad , \tag{5-1}$$

where $\phi_d{}^{L-K}$ is the phase of R(z) evaluated at some distance d fixed at 5 or more Bohr radii in front of the ion surface (i.e., $d \geq 5a_o$). At this distance the electron density differs negligibly from zero. Phase shift $\phi_d{}^{step}$, defined by eq. (2-3), is also evaluated at this same location.

The displacement of the optical surface from z = 0 is calculated using an expression similar to eq. (3-25) with $k_z = \frac{2\pi}{\lambda} \cos\theta_1$, viz.,

$$\delta = \Delta\phi / \left[2\left(\frac{2\pi}{\lambda} \cos\theta_1\right)\right] \quad . \tag{5-2}$$

The relative phase shift $\Delta\phi$ and the displacement δ as functions of the incidence angle θ_1 are shown in figure 5 for a number of wavelengths spanning the visible spectrum. Both $\Delta\phi$ and δ are negative because the apparent plane of reflection for the Lang-Kohn transition is advanced toward the light source (at negative z) by the extended electron profile. This contrasts with the results obtained for the exponential transition (which contains fewer surface electrons than the step transition).

Notice in figure 5 that although $|\Delta\phi|$ decreases with increasing wavelengths of the incident light, displacement $|\delta|$ actually increases. In accordance with eq. (5-2), δ is proportional to $\lambda\Delta\phi$ so the dependence of $\Delta\phi$ must be weaker than $1/\lambda$. This is physically interpreted to mean that at lower frequencies (larger wavelengths) the surface electrons

are more effective at shielding the interior of the metal, thereby causing $|\delta|$ to appear larger. At the same time the relative phase shift $|\Delta\phi|$ is smaller because $|\delta|$ represents a smaller fraction of the incident wavelength.

Finally observe that $|\delta|$ increases with increasing angle of incidence. At normal incidence $|\delta|$ is very much smaller than the width of the Lang-Kohn transition. However, as grazing incidence is approached the displacement $|\delta|$ of the optical surface becomes increasingly pronounced. For example, for $\theta_1 = 88°$ and $\lambda = 650$ nm, displacement $|\delta| = 11.6$ nm is more than twenty times the Lang-Kohn transition width. The physical explanation is that for the same penetration depth a grazing beam of light travels a larger distance within the transition causing the effect of the transition to become exaggerated.

6. CONCLUSIONS

The reflection of light from an optically-soft metal surface was investigated for a metal which behaves as a free-electron jellium and for which the surface transition can be described by either of two models. The first, the exponential transition, is an empirical model for which analytical results were obtained by solving Maxwell's equations for the electromagnetic fields. The second model, due to Lang and Kohn, is a semi-classical theoretical description for which the electron density within the surface transition is provided in tabular numerical form. Results in this case were obtained by means of the wave admittance formulation. For each model, the apparent plane of reflection (i.e., the optical surface of the metal) was located by comparing the phase shift upon reflection with that determined by classical Fresnel theory for an

equivalent step transition. Contrasting results were obtained for the two models. For an alkali metal the results based upon the Lang-Kohn model are more credible than those given by the exponential transition model. For other metals, not having an s-band valence structure, the exponential model may be superior. In either case, for s-polarized light reflected from a sodium surface, the computed phase shifts and optical displacements are within present observational limits. Under appropriate conditions a significant source of metrological error could result from ignoring the surface transition.

7. ACKNOWLEDGEMENT

The numerical results shown in figure 5 were computed by Jay Marchiando with the assistance of David Gilsinn.

8. REFERENCES

[1] Stratton, J. A. Electromagnetic theory. New York: McGraw-Hill Book Co.; 1941. 490-511.

[2] Kittel, C. Introduction to solid state physics. New York: John Wiley and Sons; 1966. 227-229, 244.

[3] Born, M.; Wolf, E. Principles of optics. New York: Pergamon Press; 1980. 51-70.

[4] Lieberman, A. G. An electromagnetic formulation for treating optical reflections from graded-material surfaces. Nat. Bur. Stand. (U.S.) Tech. Note 1171; 1982. 36p.

[5] Lang, N. D.; Kohn, W. Theory of metal surfaces: charge density and surface energy. Phys. Rev. B1: 4555-4568; 1970.

[6] Op. Cit. [2]. 12.

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The reflection of s-polarized light propagating in vacuum by a metal surface is examined for two descriptions of the vacuum-metal interface: the exponential surface transition and the Lang-Kohn transition. Both models treat the metal as a lossless, non-magnetic jellium material, but differ in the spatial distributions of their constituent charges. The displacement of the optical surface relative to the mechanical surface caused by the transition is evaluated for each model. Computerized results are presented for the optical displacement and phase change upon reflection from a sodium surface for the theoretically superior Lang-Kohn model. The measurement errors which result from ignoring the vacuum-metal transition become more significant as the angle of incidence is increased.

12. KEY WORDS (Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)

exponential transition; hard boundary assumption; jellium model; Lang-Kohn transition; metal surface; optical boundary; optical displacement error; optical metrology; reflection coefficient; Ricatti equation; surface transition; wave immittance

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